Quantum Monte Carlo Simulation of the Trellis Lattice Heisenberg Model for $SrCu_2O_3$ and CaV_2O_5

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We study the spin-1/2 trellis lattice Heisenberg model, a coupled spin ladder system, both by perturbation around the dimer limit and by quantum Monte Carlo simulations. We discuss the influence of the inter-ladder coupling on the spin gap and the dispersion, and present results for the temperature dependence of the uniform susceptibility. The latter was found to be parameterized well by a mean-field type scaling ansatz. Finally we discuss fits of experimental measurements on $SrCu_2O_3$ and CaV_2O_5 to our results.

KEYWORDS: spin gap, spin ladder, trellis lattice, Heisenberg antiferromagnet, $SrCu_2O_3$, CaV_2O_5, quantum Monte Carlo, scaling

§1. Introduction

Spin-1/2 ladder models[1] can describe the magnetic behavior of a variety of quasi-one dimensional materials. Examples include the cuprate materials $SrCu_2O_3$[2] and $LaCuO_{2.5}$[3] and the vanadate CaV_2O_5.[4] Spin excitations in isolated ladders have a finite energy gap, which makes them prototypical spin liquids.[1] This is of interest in relation with high temperature superconductors, since upon doping they become doped resonating-valence-bond liquids, with a spin excitation gap and dominating quasi-long range pairing correlations.

While isolated ladders are relatively easy to study and offer a variety of interesting phenomena, it is still necessary to study the inter-ladder coupling. Although the inter-ladder coupling is weak it is still necessary to obtain superconducting long range order, as observed in the ladder compound $(Sr,Ca)_{14}Cu_{24}O_{41}$.[5] Even in undoped ladder systems the weak inter-ladder coupling can play an important role. In $LaCuO_{2.5}$ an inter-ladder coupling of only one tenth of the intra-ladder coupling is sufficient to destroy the spin gap of the isolated ladder and leads to antiferromagnetic long range order.[6]

One of the authors fitted the uniform magnetic susceptibility of $SrCu_2O_3$ to that of an isolated ladder model[7] and found that the best fit is achieved with a ratio $J_\perp/J_\parallel \approx 0.5$, where $J_\perp$ is the coupling along a rung of the ladder and $J_\parallel$ that along the legs of the ladder. This result is surprising since the Cu-Cu distance across a rung is shorter than that along a leg and the rung coupling $J_\perp$ thus expected to be larger than the leg coupling. It was suggested that this apparent ratio of $J_\perp/J_\parallel \approx 0.5$ may be due to the neglect of inter-ladder interactions, although a mean-field analysis of the influence of these interactions suggested a similar ratio. In this paper we report on an investigation of the effect of the inter-ladder coupling on the magnetic susceptibility and the magnon dispersion of coupled ladders and dimers. Fits of the results to experimental measurements reveal that the inter-ladder coupling does not change the original estimates significantly and will be discussed in detail in a forthcoming publication.[8]

The susceptibility of another ladder-like compound CaV_2O_5 was fitted by Onoda and Nishiguchi to an isolated dimer model with $J_\parallel = 0$.[9] Again this result is surprising as the bond lengths are similar. We perform simulations also for this compound to estimate the strength of inter-dimer couplings.
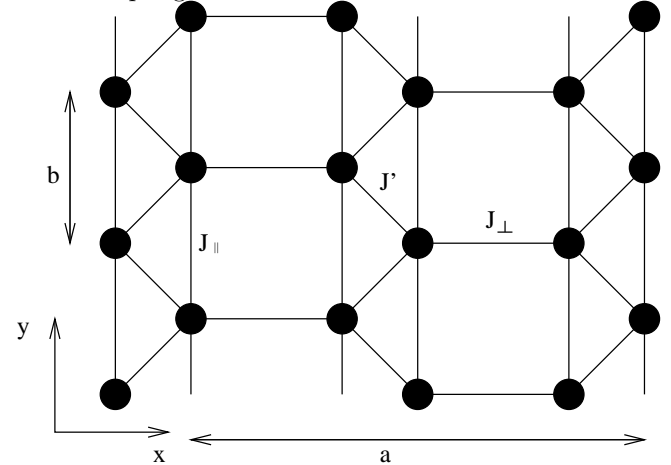

Fig. 1. The lattice structure of the trellis lattice Heisenberg model. $J_\perp$ is the exchange constant across the rung, $J_\parallel$ is that along the legs and J' is that between ladders.

Figure 1 shows the structure of the trellis lattice. It consists of spin ladders (with coupling along the legs $J_\parallel$ and across the rungs $J_\perp$, coupled by frustrating inter-ladder "zig-zag" couplings J'. The size of the lattice

used in the calculations is denoted by $N_x \times N_y$, where N_x is the number of the spin along the x-axis and N_y along the y-axis.

In $SrCu_2O_3$ and CaV_2O_5 the interactions in the ladder ($J_\perp$ and $J_\parallel$) are antiferromagnetic. The inter-ladder coupling (J') in $SrCu_2O_3$ is expected to be ferromagnetic, since it occurs via $90°$ Cu-O-Cu bonds. In CaV_2O_5 the local chemistry is more complex, and the sign of the exchange coupling J' has not been definitively established.

§2.   Spin Gap and Magnon Dispersion

In this section, we discuss the influence of the inter-ladder coupling J' on the spin gap and the magnon dispersion. Following refs.,[10,11] but extending their ideas to include the inter-ladder couplings, we start from the coupled dimer system, where $J_\perp \gg J_\parallel$, J'.

The ground state in the dimer limit ($J_\parallel = J' = 0$) consists of spin singlets on the rungs, with an energy per rung of $E_{singlet} = -\frac{3}{4}J_\perp$. Including the inter-dimer couplings perturbatively, the ground state energy per rung to second order is

$$E_0 = -\frac{3}{8}J_\perp - \frac{3}{16}\frac{J_\parallel^2}{J_\perp} - \frac{3}{32}\frac{J'^2}{J_\perp}. \quad (2.1)$$

The lowest lying excitations are spin-1 magnons, where one of the rung singlets is turned into a triplet. These magnons disperse due to inter-dimer couplings. The lowest branch is

$$\omega(k_\perp, k_\parallel) = J_\perp + \frac{3}{4}\frac{J_\parallel^2}{J_\perp} - \frac{1}{8}\frac{J'^2}{J_\perp}$$

$$+ J_\parallel \cos(k_\parallel) - \frac{1}{4}\frac{J_\parallel^2}{J_\perp}\cos(2k_\parallel)$$

$$+ (-|J'| - \frac{|J'|J'}{2J_\perp} + \frac{J_\parallel|J'|}{2J_\perp})\cos(\frac{k_\perp}{2})\cos(\frac{k_\parallel}{2})$$

$$- \frac{J'^2}{8J_\perp}(\cos(k_\perp)\cos(k_\parallel) + \cos(k_\perp) + \cos(k_\parallel))$$

$$+ \frac{J_\parallel|J'|}{2J_\perp}\cos(\frac{k_\perp}{2})\cos(\frac{3k_\parallel}{2}), \quad (2.2)$$

where we have chosen units such that the lattice parameters (see Fig. 1) are $a = b = 1$.

Minimizing $\omega(k_\perp, k_\parallel)$, we obtain for the the spin gap Δ_s in next to leading order

$$\Delta_s = \begin{cases} J_\perp - J_\parallel - \frac{J'^2}{8J_\parallel} & J_\parallel \geq |0.25J'| \\ J_\perp + J_\parallel - |J'| & J_\parallel \leq |0.25J'|. \end{cases} \quad (2.3)$$

The minimum of the dispersion is at momenta

$$k_\perp = 0 \quad (2.4)$$

$$k_\parallel = \begin{cases} 2\arccos\frac{J'}{4J_\parallel} & J_\parallel \geq |0.25J'| \\ 0 & J_\parallel \leq |0.25J'|. \end{cases} \quad (2.5)$$

We can extend above results to arbitrary ratios of $J_\parallel/J_\perp$ by replacing the second order expression of the dispersion due to $J_\parallel$

$$\epsilon^{(2)}(k_\parallel) = J_\perp + \frac{3}{4}\frac{J_\parallel^2}{J_\perp}$$

$$+ J_\parallel \cos(k_\parallel) - \frac{1}{4}\frac{J_\parallel^2}{J_\perp}\cos(2k_\parallel) \quad (2.6)$$

by the exact dispersion $\epsilon(k_\parallel)$.

Substituting the $J_\parallel$ terms in eq. (2.2) by $\epsilon(k_\parallel)$ we obtain

$$\tilde{\omega}(k_\perp, k_\parallel) = \epsilon(k_\parallel) - \frac{1}{8}\frac{J'^2}{J_\perp}$$

$$+ \left[-2|J'| - \frac{|J'|J'}{2J_\perp} + \frac{|J'|}{J_\perp}\epsilon(k_\parallel)\right]$$

$$\times \cos(\frac{k_\perp}{2})\cos(\frac{k_\parallel}{2})$$

$$- \frac{J'^2}{8J_\perp}\left[\cos(k_\perp)\cos(k_\parallel) + \cos(k_\perp)\right.$$

$$\left. + \cos(k_\parallel)\right]. \quad (2.7)$$

Expanding $\epsilon(k_\parallel)$ around the minimum at $k_\parallel = \pi$ as

$$\epsilon(\pi + k_\parallel) = \sqrt{\Delta_0^2 + v^2 k_\parallel^2}, \quad (2.8)$$

where Δ_0 is the spin gap for the spin ladder system and v is the magnon velocity we get a minimum in the dispersion at

$$\pi + (2 - \frac{\Delta_0}{J_\perp})\frac{\Delta_0^2}{v^2}(\frac{J'}{\Delta_0}), \quad (2.9)$$

and for the influence of the inter-ladder coupling J' on the spin gap

$$\Delta_s = \Delta_0\{1 - \frac{1}{2}(\frac{\Delta_0}{v})^2 C(\frac{J'}{\Delta_0})^2\}, \quad (2.10)$$

where

$$C = (2 - \frac{\Delta_0}{J_\perp})^2. \quad (2.11)$$

The change in the spin gap is small, second order in J' and with a typically small prefactor $\frac{1}{2}(\frac{\Delta_0}{v})^2 \approx 0.1$. [10,12]

We have calculated the spin gap on $(0, \pi)$ on 4×4 and 4×6 lattices by exact diagonalization using the Lanczos algorithm[13] and observed only minimal changes in the spin gap, in agreement with above arguments. These system sizes are however too small to allow quantitative comparisons.

§3.   The Uniform Susceptibility

3.1 Weakly Coupled Ladders

While the inter-ladder coupling J' ($J' \leq 0.2J_\parallel$) has negligible influence on the spin gap it still modifies the uniform magnetic susceptibility χ at intermediate and high temperatures. Using the quantum Monte Carlo loop algorithm[14,15] with improved estimators[16] we calculated the temperature dependence of χ.

We considered ladder systems with both $J_\perp/J_\parallel = 1$ and the experimentally relevant coupling $J_\perp/J_\parallel = 0.5$.[7] The inter ladder couplings were chosen to be $J'/J_\parallel = \pm 0.1, \pm 0.2, \pm 0.5$ for the isotropic ladders and $J'/J_\parallel = \pm 0.1$ and ± 0.2 for $J_\perp/J_\parallel = 0.5$.

The quantum Monte Carlo simulations suffer from a negative sign problem due to the frustrated inter-ladder couplings J'. This sign problem was alleviated a bit by

using improved estimators both for χ and for the average sign.[16] Still the sign problem restricts the QMC simulations to rather small lattices and not too low temperatures. In the temperature regime where QMC simulations were feasible a lattice size of 4×16 was sufficiently large and our results are not biased by finite size effects.
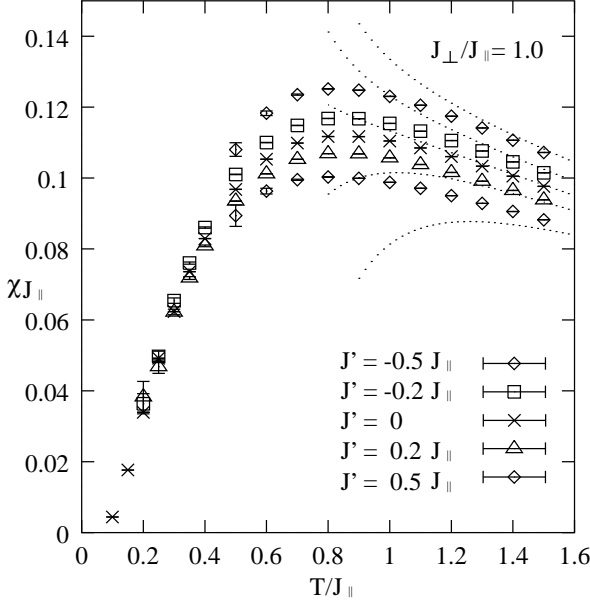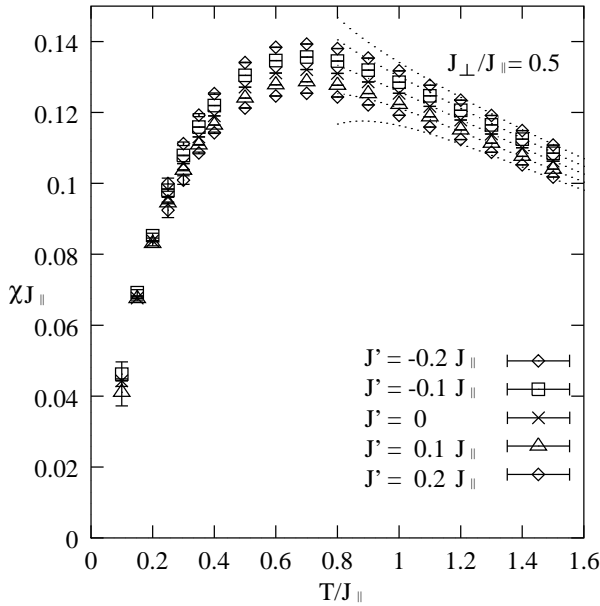

Fig. 2. Temperature dependence of the uniform susceptibility χ of the trellis lattice Heisenberg model with $J_\perp/J_\parallel = 1$ and $J'/J_\parallel = 0, \pm 0.2$ and ± 0.5. The dotted lines are fourth-order high temperature expansions.

Fig. 3. Temperature dependence of the uniform susceptibility χ of the trellis lattice Heisenberg model with $J_\perp/J_\parallel = 0.5$ and $J'/J_\parallel = 0, \pm 0.2$. The dotted lines are fourth-order high temperature expansions.

Our results are shown in Figs. 2 and 3 together with

a fourth-order high temperature expansion

$$
\begin{aligned}
\chi(T) = {} & \frac{1}{4T} - \frac{1}{16}(J_\perp + 2J_\parallel + 2J')(\frac{1}{T})^2 \\
& + \frac{1}{64}(4J_\perp J_\parallel + 4J_\perp J' + 8J_\parallel J' - J_\perp{}^2)(\frac{1}{T})^3 \\
& - \frac{1}{768}(-J_\perp{}^3 - 8J_\parallel{}^3 - 8J'^3 - 6J_\perp{}^2 J_\parallel \\
& - 6J_\perp{}^2 J' + 24J_\parallel{}^2 J' + 12J_\perp J_\parallel{}^2 + 12J_\perp J'^2 \\
& + 6J_\parallel J'^2 + 72J_\perp J_\parallel J')(\frac{1}{T})^4.
\end{aligned} \tag{3.1}
$$

Fitting magnetic susceptibility measurements on spin ladder materials is much simplified if approximate analytic expressions for the magnetic susceptibility are available. The single ladder susceptibilities have been parameterized before.[7, 8, 12, 17]

To extend these parameterizations to include the inter-ladder coupling we follow a mean field-type scaling ansatz (MFTS) of ref.[7] and scale the trellis lattice susceptibility to the susceptibility $\chi_l(J_\perp/J_\parallel)$ of a single ladder:

$$
\chi(\frac{J_\perp}{J_\parallel}, \frac{J'}{J_\parallel}) = \frac{\chi_l(\frac{J_\perp}{J_\parallel})}{1 + f(\frac{J'}{J_\parallel})\chi_l(\frac{J_\perp}{J_\parallel})J_\parallel}, \tag{3.2}
$$

with

$$
f(\frac{J'}{J_\parallel}) = A\frac{J'}{J_\parallel} + B(\frac{J'}{J_\parallel})^2, \tag{3.3}
$$

and A and B as temperature independent constants. At low temperatures this ansatz gives the same gapped behavior as a single ladder, which is reasonable since the gap is not changed much. Fixing $A = 2$ also recovers the correct high temperature Curie-Weiss law. By fitting our QMC results in the temperature range $1.0 \leq T/J_\perp \leq 1.5$ to eq. (3.2) we obtain

$$
B = 0.3436(3). \tag{3.4}
$$

As a check of the quality of the MFTS ansatz we compare, in Fig. 4, the single ladder susceptibilities obtained from the QMC data of coupled ladders by inverting eq. (3.2). We find that the ansatz is useful for the whole temperature range as long as the inter-ladder coupling J' is small.

3.2 Coupled Dimers

We can repeat a similar analysis for weakly coupled dimers instead of weakly coupled ladders. We considered leg couplings of $J_\parallel/J_\perp = 0$, 0.1 and 0.2 and inter-ladder couplings $J'/J_\perp = 0$, ± 0.1 and ± 0.2 on systems of size 32×16, where finite size effects are again negligible. In Fig. 5 we show the uniform susceptibility for $J_\parallel/J_\perp = 0.1$.

We again make use of a MFTS ansatz to obtain an approximate analytic expression for the susceptibility

$$
\chi(\frac{J_\parallel}{J_\perp}, \frac{J'}{J_\perp}) = \frac{\chi_l(\frac{J_\parallel}{J_\perp})}{1 + \tilde{f}(\frac{J'}{J_\perp})\chi_l(\frac{J_\parallel}{J_\perp})J_\perp}, \tag{3.5}
$$
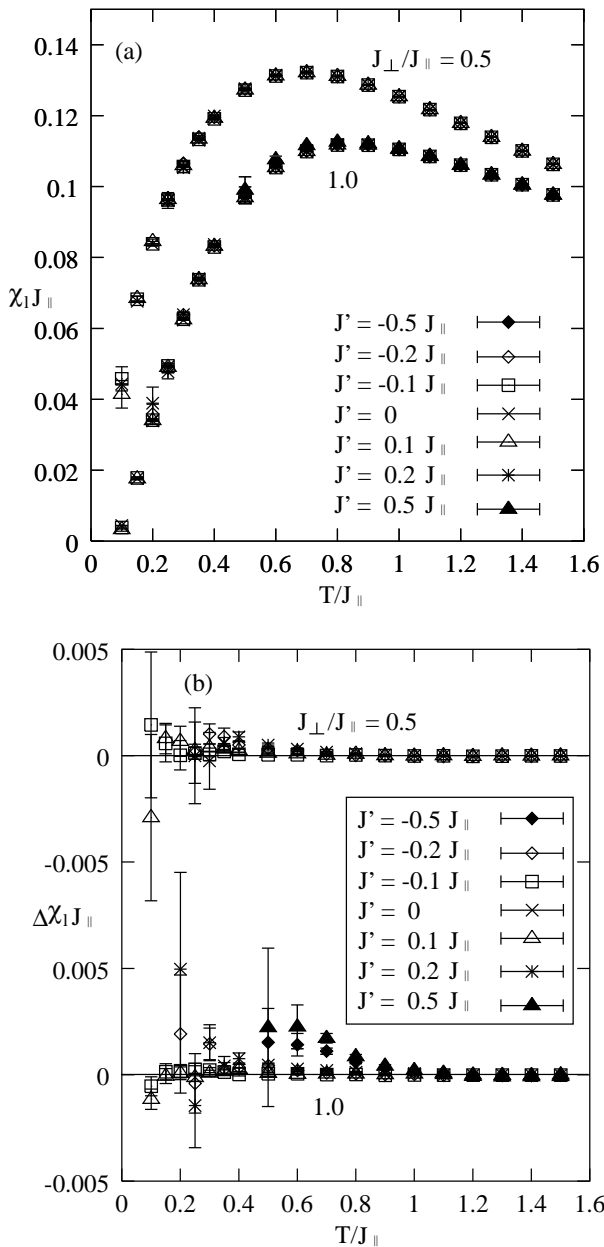
Fig. 4.   (a) Scaling plot of weakly coupled ladders, (b) differences between the scaled and actual susceptibilities.

with

$$\tilde{f}(\frac{J'}{J_\perp}) = \tilde{A}\frac{J'}{J_\perp} + \tilde{B}(\frac{J'}{J_\perp})^2. \tag{3.6}$$

and $\tilde{A}$ and $\tilde{B}$ temperature independent constants. The high temperature expansion again fixes $\tilde{A} = 2$, and fits to the QMC susceptibility data in the temperature range $1.0 \le T/J_\perp \le 1.5$ give

$$\tilde{B} = 0.6940(3). \tag{3.7}$$

Scaling plots (Figure 6) show that the agreement is not as good as in the coupled ladder case. This is not very surprising, since, as can be seen in eq.(2.3) and eq.(2.10), the spin gap of weakly coupled dimers depends more on the inter-dimer couplings than that of
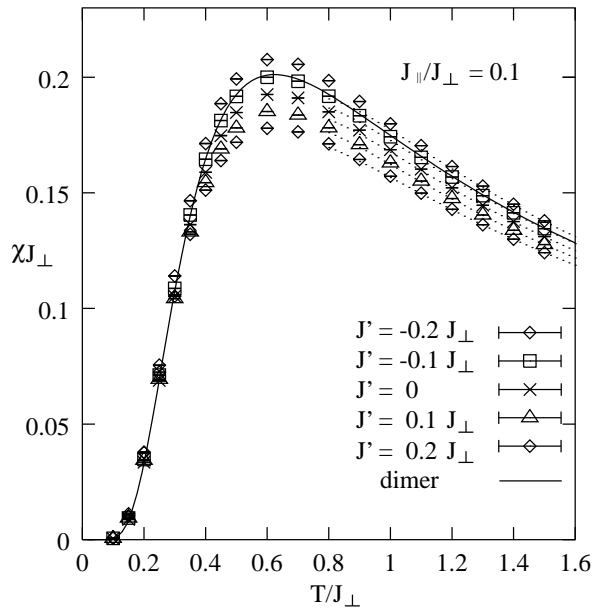

Fig. 5.   Temperature dependence of the magnetic susceptibility of the trellis lattice Heisenberg model in the regime of weakly coupled dimer. The coupling ratios are $J_\parallel/J_\perp = 0.1$ and $J'/J_\perp = \pm 0.1$ and ± 0.2. The dotted lines are again the fourth order high temperature expansions.

weakly coupled ladders. Still the MFTS ansatz gives a reasonable analytic parameterization of the susceptibilities of weakly coupled dimers.

§4.   Comparison with Experiments

In this section we wish to briefly compare our QMC results with experimental measurements of the susceptibility of $SrCu_2O_3$ and CaV_2O_5. A forthcoming publication[8] will present detailed fits and comparisons.

4.1 $SrCu_2O_3$

With regard to fits of the experimental susceptibility measurements on $SrCu_2O_3$ our main result is that the inclusion of inter-ladder couplings does not modify the previous estimate $J_\perp/J_\parallel \approx 0.5$[7] considerably. We cannot determine the value of J' from these fits since in the experimentally accessible temperature range $T < 650$K the dependence of the susceptibility on J' is weak.

4.2 CaV_2O_5

It was proposed previously that the magnetic susceptibility of CaV_2O_5 can be fit by that of dimers.[9] We have performed a series of fits on new experimental data of Isobe and Ueda.[18]

One problem is that the samples are not pure CaV_2O_5 but contain a few percent CaV_3O_7. By X-ray structural analysis Isobe and Ueda determined that the current sample contains 4.1% CaV_3O_7. By subtracting the separately measured susceptibility of CaV_3O_7[18] we obtained experimental data for pure CaV_2O_5.

The main result of our fits is that the inter-dimer couplings $J_\parallel$ and J' are both much smaller than $J_\perp$, and we can give some constraints.

To determine the spin gap Δ we fitted the low temperature experimental data to the expression for a spin ladder at $T \ll \Delta$[12)]

$$\chi(T) = \frac{C}{T - \Theta} + \chi_0 + \frac{a}{\sqrt{T}} \exp(-\Delta/T), \qquad (4.1)$$

where the g-factor of Vanadium was determined to be $g = 1.96$ by ESR measurements.[9, 18)] The fit parameters were $C = 1.6 \times 10^{-3}$ cm^{3}K/mol V, $\Theta = -8$K and $\chi_0 = 1.23 \times 10^{-5}$ cm^3/mol V. We use this fit to subtract the first term, which is a Curie-Weiss term due to paramagnetic impurities, and the second, temperature independent, constant contribution.

Next we fit the low temperature data ($T < 200$K) to QMC results for an isolated ladder for $J_\parallel/J_\perp = \pm 0.1$, ± 0.2 and ± 0.3. This fit is reasonable because we will see later, that we are in the ladder case $|J'| \ll J_\parallel$, where the influence of J' on the low temperature susceptibility is negligible. In all cases we found $J_\perp \approx 670$K.
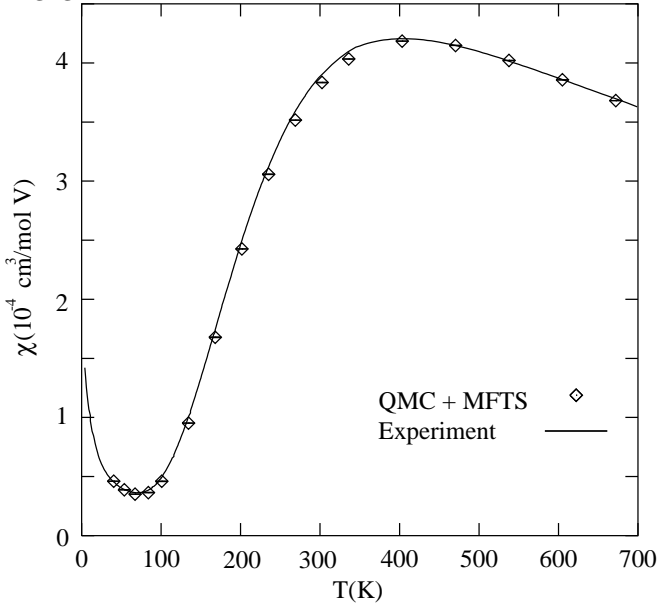

Fig. 7. Fit of the uniform magnetic susceptibility for CaV$_2$O$_5$, assuming $J_\parallel/J_\perp = 0.1$. The fit parameters are $J_\perp = 672$K, $J_\parallel = 67$K and $J' = 45$K

Next, for each of the above ratios of $J_\parallel/J_\perp$ with $J'/J_\perp = 0$ we use the MFTS eq. (3.5) to fit the susceptibility in the range 200K $< T < 700$K and determine J'. Good fits are obtained only for $J_\parallel/J_\perp = 0.1$ and 0.2, but not for $J_\parallel/J_\perp \leq 0$ or $J_\parallel/J_\perp = 0.3$. Assuming $J_\parallel/J_\perp = 0.1$ we obtained $J_\perp \approx 670$K, $J_\parallel \approx 67$K and $J' \approx 45$K. This fit is shown in Fig. 7. For the other ratio $J_\parallel/J_\perp = 0.2$ we got an equally good fit with $J_\perp \approx 665$K, $J_\parallel \approx 135$K and a ferromagnetic $J' \approx -25$K.

Given the good quality of both fits it is hard to determine the exact values of the couplings $J_\parallel$ and J' from fits to the uniform susceptibility alone. But we can give some estimates and constraints: $J_\perp \approx 670$K, 0K $< J_\parallel < 200$K and $J' + J_\parallel \approx 110$K. The dispersion relation of the magnons however depends sensitively on the ratio $J_\parallel/J'$ and could offer a way to determine these couplings more precisely.

§5.   Conclusions

We have studied the trellis lattice Heisenberg model by quantum Monte Carlo and a perturbation expansion around the dimer limit. We confirmed that for weakly coupled ladders the influence of the frustrated inter-ladder coupling on the spin gap is small.

We calculated the uniform susceptibility by QMC simulations and found that a mean field-type scaling ansatz gives a reasonable analytic parameterization of its temperature dependence in the whole temperature range.

This parameterization was in turn be used to fit experimental measurements on the compounds SrCu$_2$O$_3$ and CaV$_2$O$_5$. For SrCu$_2$O$_3$ it was found that the inter-ladder coupling does not significantly modify the previous estimate of the intra-ladder coupling ratio $J_\perp/J_\parallel \approx 0.5$.[7)] CaV$_2O_5$ was confirmed to be a weakly coupled dimer system with inter-dimer couplings about an order of magnitude smaller than the intra-dimer coupling.

1) E. Dagotto and T.M. Rice, Science **271** (1996) 618.
2) M. Azuma, Z. Hiroi, M. Takano, K. Ishida and Y. Kitaoka, Phys. Rev. Lett. **73** (1994) 3463.
3) Z. Hiroi and M. Takano, Nature **377**, 41 (1995).
4) H. Iwase, M. Isobe, Y. Ueda and H.Yasuoka, J. Phys. Soc. Jpn. **65** (1996) 2397.
5) M. Uehara *et al.*, J. Phys. Soc. Jpn. **65**. 2764 (1996).
6) B. Normand and T.M. Rice, Phys. Rev. B **54**, 7180 (1996); M. Troyer, M.E. Zhitomirsky and K. Ueda, Phys. Rev. B **55**, R6117 (1997).
7) D. C. Johnston, Phys. Rev. B **54** (1996) 13009.
8) D.C. Johnston *et al.*, in preparation.
9) M. Onoda and N. Nishiguchi, J. Solid State Chem. **127** (1996) 359.
10) T. Barnes, E. Dagotto, J. Riera and E.S. Swanson, Phys. Rev. B **47**, 3196 (1993).
11) M. Reigrotzki, H. Tsunetsugu and T.M. Rice, J. Phys. Cond. Matt. **6**, 9235 (1994).
12) M. Troyer, H. Tsunetsugu and D. Würtz, Phys. Rev. B **50**, (1994) 13515.
13) C. Lanczos, J. Res. Natl. Bur. Stand. **45**, 225 (1950).
14) H.G. Evertz, G. Lana and M. Marcu, Phys. rev. Lett. **70**, (1993) 875.
15) H.G. Evertz, cond-mat/9707221, to be published in *"Numerical Methods for Lattice Quantum Many-Body Problems"*, ed. D.J. Scalapino, Addison Wesley Longman, Frontiers in Physics.

16) B. Ammon, H.G. Evertz, N. Kawashima, M. Troyer and B.
    Frischmuth, cond-mat/9711022.
17) T. Barnes and J. Riera, Phys. Rev. B **50**, 6817 (1994).
18) M. Isobe and Y. Ueda, unpublished data.
19) M. Troyer, B. Ammon and E. Heeb, *Parallel object oriented
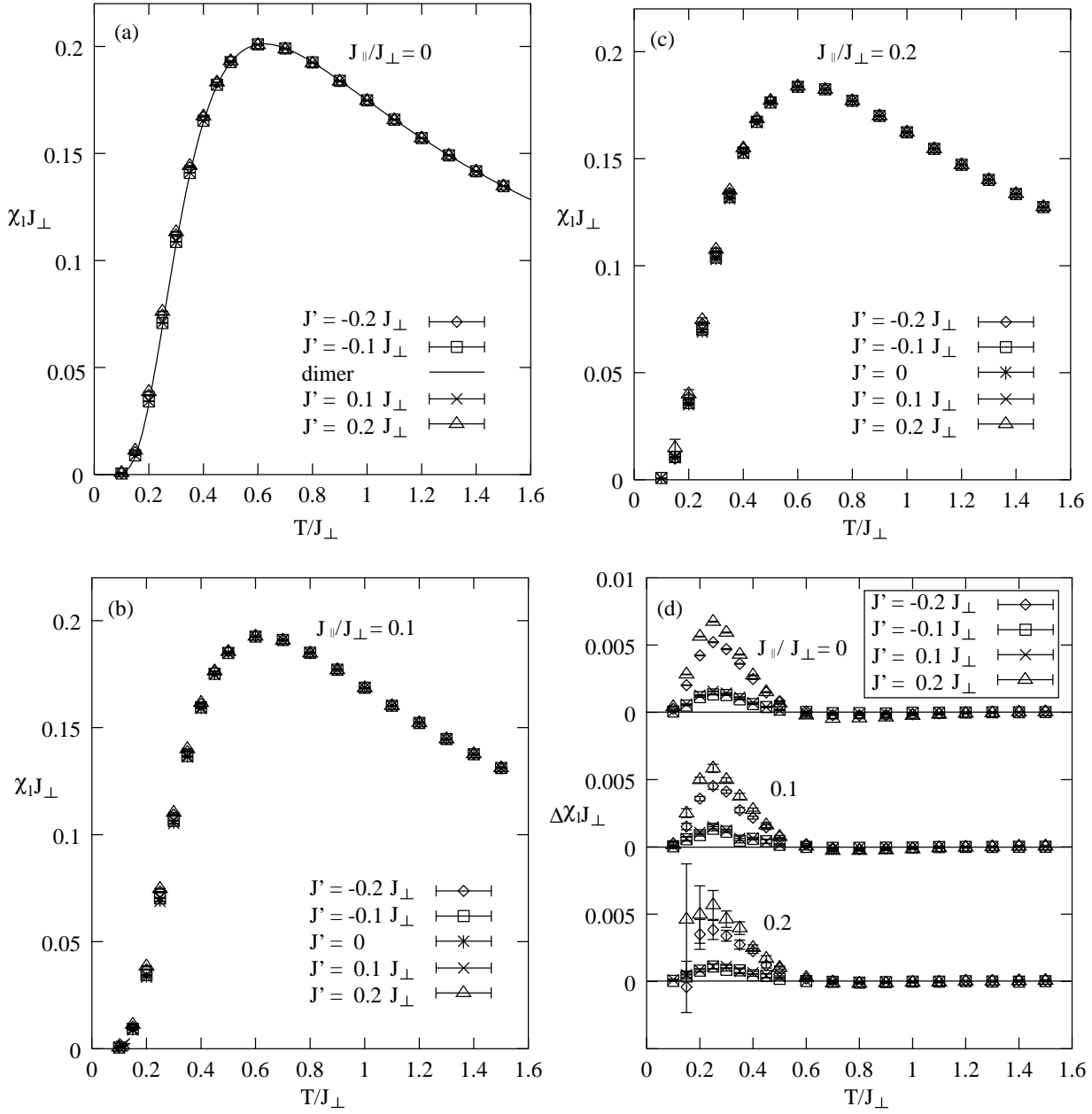   Monte Carlo simulations*, preprint.

Fig. 6.   Scaling plots for the weakly coupled dimer regime for couplings (a) $J_\parallel/J_\perp = 0$, (b) $J_\parallel/J_\perp = 0.1$ and (c) $J_\parallel/J_\perp = 0.2$. (d) shows the differences between the scaling curves